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(71) Applicant: 000003300

Tosoh Corporation

4560, Kaisei-cho, Shin Nanyo-shi, Yamaguchi-ken

(72) Inventor:

Masayuki Yamaguchi

10-3, Betsumei 3-chome, Yokkaichi-shi, Mie-ken

(72) Inventor:

Kenichi Suzuki

5-1, Betsumei 3-chome, Yokkaichi-shi, Mie-ken

(72) Hiroshi Miyata

129, Hazuotsu, Yokkaichi-shi, Mie-ken,

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Propylene Resin Composition

(57) Abstract

[Problem]

To provide a propylene resin composition suitable to fabrication such as blow molding, vacuum forming, pressure forming, film forming, lamination molding, fiber spinning, and expansion molding.

[Means for Resolution]

A propylene resin composition characterized in that it comprises a crosslinked product of an aromatic vinyl/hydrogenated diene block copolymer satisfying specific characteristics, and a propylene resin, and satisfies following properties (c) and (d):

(c) the propylene resin composition can be drawn so as to satisfy the relationship: $d \leq 0.25 \times D$ wherein d is a diameter of a strand extruded from a circular die having an inside diameter D.

(d) a ratio of the maximum values of elongational viscosity to shear viscosity both of which are measured at the same temperature and the same strain rate satisfies the relationship: elongational viscosity/shear viscosity ≥ 4 , is used.

Claim

[Claim 1]

A propylene resin composition characterized in that it comprises a crosslinked product of an aromatic vinyl/hydrogenated diene block copolymer satisfying characteristics (a) and (b) described below, and a propylene resin, and satisfies properties (c) and (d) described below:

(a) the aromatic vinyl/hydrogenated diene block copolymer is a block copolymer comprising (A) a polymer block unit mainly comprising an aromatic vinyl compound, (B) a hydrogenated conjugated diene block unit having 1,2-bond units of the conjugated diene being at least 65 mol%, and at least 90% of double bond units originated from the conjugated diene being hydrogenated and saturated, or a random copolymer block unit comprising the hydrogenated conjugated diene block unit and an aromatic vinyl compound unit, and/or (C) a taper block unit comprising an aromatic vinyl compound and a conjugated diene in which double bond units originated from the conjugated diene being hydrogenated, and the aromatic vinyl compound gradually increases, being of an (A)-(B), (A)-(B)-(A) or (A)-(B)-(C) type,

(b) a ratio of units originated from the aromatic vinyl compound to those from the conjugated diene

satisfies the relationship: aromatic vinyl compound/conjugated diene = 3/97 to 50/50 (weight ratio),

(c) the propylene resin composition can be drawn so as to satisfy the relationship: $d \leq 0.25 \times D$ wherein d is the diameter of a strand extruded from a circular die having an inside diameter D, and

(d) a ratio of the maximum values of elongational viscosity to shear viscosity both of which are measured at the same temperature and the same strain rate satisfies the relationship: elongational viscosity/shear viscosity ≥ 4 .

[Detailed Description of the Invention]

[0001]

[Technical Field to which the Invention Belongs]

The invention relates to a propylene resin composition excellent in molding processability.

[0002]

[Prior Art]

A propylene resin has been well known as a commonly used polymer material excellent in heat resistance. It is known however that when the propylene resin is subjected to blow molding, vacuum forming, pressure forming, film forming, lamination molding, fiber spinning, or expansion molding, it involves problems such as uneven thickness, neck-in, surging, resonance, and breakage of foams. Many

of the problems of molding defects are related to elongational viscosity, and it is known generally that the moldability is improved when the elongational viscosity is sufficiently greater than shear viscosity at the same temperature and the same strain rate, (M. Shinohara, Nihon Reoroji Gakkaishi (Journal of the Society of Rheology, Japan) Vol.19, p. 118 (1991)). However, the elongational viscosity of the polypropylene resin is usually at most about three times the shear viscosity at the same strain rate.

[0003]

Then, for increasing the elongational viscosity of the polypropylene resin, an approach of widening a molecular weight distribution of the propylene resin or adding a branched low density polyethylene (LDPE) is performed.

[0004]

Moreover, a polypropylene having a long chain branch was developed recently and put into the market (*Plastic Engineering* p. 82, March '91).

[0005]

[Problems that the Invention is to Solve]

However, in the method of widening the molecular weight distribution of the propylene resin, it is difficult to increase the ratio of elongational

viscosity/shear viscosity of the propylene resin at the same temperature and the same strain rate to greater than 3, so that the effect of improving the molding processability is insufficient.

[0006]

On the other hand, a polypropylene having a branched long chain is inferior in melt-drawing property, therefore, molding defects occurred frequently.

[0007]

Then, the present invention provides a propylene resin composition suitable to blow molding, vacuum forming, pressure forming, film forming, lamination molding, fiber spinning, expansion molding, etc. which solved the problems described above.

[0008]

[Means for Solving the Problem]

As a result of intensive studies to solve the problems, the present inventors have found that a propylene resin composition having specific properties exhibits excellent processability, and have accomplished the present invention.

[0009]

That is, the invention relates to a propylene resin composition characterized in that it comprises a crosslinked product of an aromatic vinyl/hydrogenated

diene block copolymer satisfying characteristics (a) and (b) described below, and a propylene resin, and satisfies properties (c) and (d) described below.

[0010]

(a) the aromatic vinyl/hydrogenated diene block copolymer is a block copolymer comprising (A) a polymer block unit mainly comprising an aromatic vinyl compound, (B) a hydrogenated conjugated diene block unit having 1,2-bond units of the conjugated diene being at least 65 mol%, and at least 90% of double bond units originated from the conjugated diene being hydrogenated and saturated, or a random copolymer block unit comprising the hydrogenated conjugated diene block unit and an aromatic vinyl compound unit, and/or (C) a taper block unit comprising an aromatic vinyl compound and a conjugated diene in which double bond units originated from the conjugated diene being hydrogenated, and the aromatic vinyl compound gradually increases, being of an (A)-(B), (A)-(B)-(A) or (A)-(B)-(C) type.

[0011]

(b) a ratio of units originated from the aromatic vinyl compound to those from the conjugated diene satisfies the relationship: aromatic vinyl compound/conjugated diene = 3/97 to 50/50 (weight ratio).

[0012]

(c) the propylene resin composition can be drawn so as to satisfy the relationship: $d \leq 0.25 \times D$ wherein d is the diameter of a strand extruded from a circular die having an inside diameter D.

[0013]

(d) a ratio of the maximum values of elongational viscosity to shear viscosity both of which are measured at the same temperature and the same strain rate satisfies the relationship: elongational viscosity/shear viscosity ≥ 4 .

[0014]

The present invention will be described specifically below.

[0015]

The propylene resin constituting the propylene resin composition of the invention may be a generally known crystalline propylene resin, and includes, for example, a propylene homopolymer, a propylene/ethylene random copolymer having an ethylene content of from 0.5 to 12% by weight, a propylene/ethylene/ α -olefin terpolymer having an ethylene content of from 0.5 to 12% by weight, and a content of an α -olefin such as 1-butene of from 0.5 to 20% by weight, a high impact polyethylene having an ethylene content of from 1 to 60% by weight, a polypropylene having a branched long chain introduced therein, and a

syndiotactic polypropylene. They are used alone or in combination of two or more of them. It is preferred that the propylene resin has a melt flow rate (hereinafter referred to as MFR) at 230°C under the load of 2.16 kg of 0.01 to 100 g/10 min.

[0016]

It is preferred that an aromatic vinyl/hydrogenated diene block copolymer to be used for obtaining the crosslinked product of an aromatic vinyl/hydrogenated diene block copolymer constituting the propylene resin of the invention is preferably a block copolymer comprising (A) a polymer block unit mainly comprising an aromatic vinyl compound, (B) a hydrogenated conjugated diene block unit having 1,2-bond units of the conjugated diene being at least 65 mol%, and at least 90% of double bond units originated from the conjugated diene being hydrogenated and saturated, or a random copolymer block unit comprising the hydrogenated conjugated diene block unit and an aromatic vinyl compound unit, and/or (C) a taper block unit comprising an aromatic vinyl compound and a conjugated diene in which double bond units originated from the conjugated diene being hydrogenated, and the aromatic vinyl compound gradually increases, being of an (A)-(B), (A)-(B)-(A) or (A)-(B)-(C) type, because a resulting propylene resin composition is excellent in

molding processability, weathering resistance and heat resistance, and it is also preferred that a ratio of units originated from the aromatic vinyl compound to those from the conjugated diene satisfies the relationship: aromatic vinyl compound/conjugated diene = 3/97 to 50/50 (weight ratio), because excellent handleability of the aromatic vinyl/hydrogenated diene block copolymer is provided, and a resulting propylene resin composition is excellent in flexibility and molding processability.

[0017]

The aromatic vinyl compound herein includes, for example, styrene, t-butyl styrene, α -methyl styrene, p-methyl styrene, divinyl benzene, 1,1-diphenyl styrene, N,N-diethyl-p-aminoethyl styrene and vinyl pyridine, with styrene and α -methyl styrene being particularly preferred. The conjugated diene compound includes, for example, 1,3-butadiene, 1,3-pentadiene, 2-methyl-1,3-pentadiene, 1,3-hexadiene, 4,5-diethyl-1,3-octadiene, 3-butyl-1,3-octadiene, and chloroprene, with 1,3-butadiene and isoprene being particularly preferred.

[0018]

The aromatic vinyl/hydrogenated diene block copolymer preferably has a number average molecular weight of from 30,000 to 1,000,000, particularly, from 50,000 to 600,000 in terms of polystyrene, because crosslinking can

be performed easily, and excellent handleability is provided.

[0019]

Such aromatic vinyl/hydrogenated diene block copolymer is obtained by a method disclosed in JP-A-3-72512, namely, a method of performing living anion polymerization of the block unit (A), the block unit (B) and if required the taper block unit (C) or the block unit (A) in an organic solvent using an organic alkali metal compound as an initiator to obtain a block copolymer, and further hydrogenating the obtained block copolymer.

[0020]

A method for crosslinking the aromatic vinyl/hydrogenated diene block copolymer is not particularly limited, and includes, for example, a method of performing crosslinking with electron rays, a peroxide, or the like.

[0021]

When performing crosslinking with electron rays, the irradiation dose is preferably 10 kGray or more because moldability of the resulting propylene resin composition is excellent.

[0022]

Further, when performing thermal crosslinking with a peroxide, the peroxide includes, for example,

ketoneperoxides such as methylethyl ketone peroxide, cyclohexanone peroxide, 3,3,5-trimethylhexanone peroxide, methylcyclohexanone peroxide, methylacetacetate peroxide, and acetylacetone peroxide; peroxyketals such as 1,1-bis(t-butyl peroxy)-3,3,5-trimethyl cyclohexane, 1,1-bis(t-butyl peroxy)cyclohexane, 2,2-bis(t-butyl peroxy)octane, 2,2-bis(t-butyl peroxy)butane, and n-butyl-4,4-bis(t-butyl peroxy)hexane; hydroperoxides such as t-butylhydroperoxide, cumene hydroperoxide, di-isopropylbenzene hydroperoxide, p-menthane hydroperoxide, 2,5-dimethyl hexane-2,5-di-hydroperoxide, and 1,1,3,3-tetramethyl butyl hydroperoxide; dialkyl peroxides such as di-t-butyl peroxide, t-butylcumylperoxide, dicumyl peroxide, α , α' -bis(t-butyl peroxy-m-isopropyl)benzene, and 2,5-dimethyl-2,5-di-(t-butyl peroxy)hexine-3; diacylperoxides such as acetyl peroxide, isobutyl peroxide, octanoyl peroxide, decanoyl peroxide, lauroyl peroxide, 3,3,5-trimethyl hexanoyl peroxide, peroxide succinate, benzoyl peroxide, and 2,4-dichlorobenzoyl peroxide; peroxydicarbonates such as di-isopropyl peroxydicarbonate, di-2-ethylhexyl peroxydicarbonate, di-n-propyl peroxydicarbonate, bis(4-t-butyl cyclohexyl)peroxydicarbonate, di-2-ethoxyethyl peroxydicarbonate, di-methoxyisopropyl peroxydicarbonate, di-(3-methyl-3-methoxybutyl)peroxydicarbonate, and diallyl

peroxydicarbonate; and proxyesters such as t-butyl peroxy acetate, t-butyl peroxy isobutyrate, t-butyl peroxy laurate, t-butyl peroxy benzoate, t-butyl peroxy benzoate, di-t-butyl peroxy isophthalate, 2,5-dimethyl-2,5-di-(benzoyl peroxy)hexane, and t-butyl peroxide isopropyl carbonate, and they may be used alone, or in combination of two or more of them.

[0023]

Further, when performing crosslinking with a peroxide, a crosslinking promoter and a co-crosslinking agent can be used in combination. The crosslinking promoter includes, for example, guanidine derivatives such as N,N-diphenyl guanidine, N,N-di-(o-tolyl)guanidine, and N,N-o-tolylguanidine; thioureas such as N,N-dibutyl thiourea, N,N'-diethyl thiourea, dilauryl thiourea, 2-mercaptoimidazoline, trimethyl thiourea, and tetramethyl thiourea; xanthogenates such as zinc dibutyl xanthogenate, sodium isopropyl xanthogenate, and zinc isopropyl xanthogenate; dithiocarbamates such as zinc dimethyl dithiocarbamate, zinc diethyl dithiocarbamate, zinc dibutyl dithiocarbamate, zinc ethylphenyl dithiocarbamate, zinc N-pentamethylene dithiocarbamate, zinc dibenzyl dithiocarbamate, sodium dimethyl dithiocarbamate, sodium diethyl dithiocarbamate, sodium dibutyl dithiocarbamate, copper dimethyl dithiocarbamate, tellurium diethyl

dithiocarbamate, piperidinium pentamethylene dithiocarbamate, pipecoline piperidimethyl dithiocarbamate, and iron dimethyl dithiocarbamate; aldehyde ammonia-based compounds such as hexamethylene tetramine, acetoaldehydeaniline, and butyl aldehydeaniline; thiazole-based compounds such as mercaptobenzothiazole, a sodium salt of mercaptobenzothiazole, dibenzothiazyldisulfide, 2-(4-morpholino dithio)benzothiazole, and 2-(2,4-dinitrophenyl)mercaptobenzothiazole; thiuram sulfides such as tetramethyl thiurum monosulfide, tetraethyl thiurum monosulfide, tetramethyl thiurum disulfide, tetraethyl thiurum disulfide, tetrabutyl thiurum disulfide, and dipentamethylene thiurum tetrasulfide; thiazole-based compounds such as mercaptobenzothiazole, dibenzothiazyldisulfide, and zinc 2-mercaptobenzothiazole; and sulfenamide compounds such as N-cyclohexyl-2-benzothiazole sulfenamide, N-oxydiethylene-2-benzothiazole sulfenamide, and N-t-butyl benzothiazole sulfenamide. Those crosslinking promoters may be used alone or in combination of two or more of them.

[0024]

The co-crosslinking agent includes, for example, p-quinonedioxime, p,p-dibenzoylquinonedioxime, N-methyl-N'-4-dinitrosoaniline, dinitrosobenzene, lauryl methacrylate, ethylene glycol dimethacrylate, triethylene glycol

dimethacrylate, tetraethylene glycol dimethacrylate, polyethylene glycol dimethacrylate, trimethylol propane trimethacrylate, diarylfumarate, diarylphtalate, tetraaryloxyethane, triarylcyanurate, arylmethacrylate, maleimide, phenylmaleimide, N,N'-m-phenylenebismaleimide, maleic anhydride, itaconic acid, divinylbenzene, diaryl melamine, diphenyl guanidine, divinyl adipate, vinyl toluene, 1,2-polybutadiene, liquid styrene-butadiene copolymer rubber, dipentamethylene thiuram pentasulfide, mercaptobenzthiazole, and sulfur. They may be used alone or in combination of two or more of them.

[0025]

Besides, a crosslinking aid, for example, zinc white, active zinc white, surface-treated zinc white, zinc carbonate, litharge or magnesium oxide, and a dispersing agent and the like may be used in combination.

[0026]

For improving the moldability of the propylene resin composition of the invention, it is preferred that MFR (MFR_a) of the aromatic vinyl-hydrogenated diene block copolymer before subjected to crosslinking and MFR (MFR_b) thereof after subjected to crosslinking satisfy the relationship: MFR_a/MFR_b > 1, particularly, MFR_a/MFR_b > 5. The MFR is a value measured according to JIS K6758 under the conditions of a temperature of 230°C and a load of

2.16 kg.

[0027]

The propylene resin composition of the present invention is one which can be drawn so as to satisfy the relationship: $d \leq 0.25 \times D$, preferably, $d \leq 0.20 \times D$ wherein d is the diameter of a strand extruded from a circular die having an inside diameter D when evaluating a melt-drawing property. The resin composition, when it cannot be drawn so as to satisfy the relationship: $d \leq 0.25 \times D$, is inferior in melt-drawing property, and therefore, it is poor in moldability when subjected to molding process, so that it cannot be drawn to a high level to result in formation of products with poor appearance.

[0028]

As a method of measuring melt-drawing property, generally known methods can be used, including, for example, a method of using an actual molding machine, and a method of using a capillary rheometer. The measurement is performed preferably at a temperature of from 170 to 250°C which is a general temperature range as a measurement condition of polypropylene resins. There is no particular limitation on the inside diameter D and the length L of the die used for the measurement. The measurement is preferably performed using a die having an

inside diameter D of 0.1 to 5 mm and the length L of 0.1 to 100 mm, at an extrusion rate of the strand of 0.02 to 2 m/min and at a residence time of 3 to 30 min, because the measurement is facilitated. In the present invention, the diameter d of the strand was measured when the measurement was performed at the drawable maximum take-off speed under the conditions of 190°C, the cylinder descending speed of 10 mm/min, and a residence time of 6 min and was determined by a capillary type rheometer, using a die having L/D of 2.95/1 (mm).

[0029]

The propylene resin composition of the invention has a ratio of the maximum values of elongational viscosity to shear viscosity measured at the same temperature and the same strain rate satisfying the relationship: elongational viscosity/shear viscosity \geq 4, preferably, elongational viscosity/shear viscosity \geq 4.5. When the ratio of elongational viscosity/shear viscosity is less than 4, resulting propylene resin compositions tend to suffer from molding defects during the molding process, such as uneven thickness, neck-in, resonance, or surging to result in those with poor molding processability. The method of the measurement is performed such that the temperature is preferably 170 to 250°C, which is an actual processing temperature, and the strain rate is preferably 0.01 to 1

sec^{-1} . The measurement of the shear viscosity is preferably performed using a cone-plate type viscometer, and the measurement method thereof is, for example, the method described in Ferry, *Viscoelastic Properties of Polymers*, Third edition, Wiley, New York (1980). Further, the measurement method of the elongational viscosity is, for example, the method described in O. Ishizuka, et al., *Polymer*, Vol. 21, p.164(1980). The measurement of the elongational viscosity in the present invention was performed using a Meissner type elongational viscometer, at a temperature of 190°C, and at a strain rate of $0.1 \pm 0.05 \text{ sec}^{-1}$. Further, the shear viscosity was measured using a cone-plate type viscometer at a temperature of 190°C, and thus the shear viscosity at a strain rate of 0.1 sec^{-1} was determined.

[0030]

For reducing the occurrence of uneven thickness and providing excellent molding processability, the propylene resin of the present invention has a melt tension of preferably 3 g or more. The measurement condition of the melt tension is the same as the measurement condition of the melt drawing property.

[0031]

Further, for forming a resin composition excellent in processability such as, in particular, melt drawing

property, the propylene resin composition of the present invention has a flow activation energy of preferably from 35 to 45 kJ/mol. The measurement method of the flow activation energy can be the method described in, for example, *Koza (Course) Rheology*, edited by Rheology Society, published by Koubunsi Kanko Kai, etc. That is, the flow activation energy can be determined by measuring a dynamic viscoelasticity of the polypropylene resin composition of the present invention at least at two optional temperatures of from 170°C to the decomposition temperature of the propylene resin composition.

[0032]

A melt flow rate (measured under 230°C, 2.16 kg load) of a propylene-based resin composition of the present invention is preferably within the range of 0.01 to 100 g/10 min, because if the melt flow rate is within the range, good processability can be obtained.

[0033]

In the propylene resin composition of the invention, the blending ratio of the propylene resin to the crosslinked product of the aromatic vinyl/hydrogenated diene block copolymer satisfies, though not particularly restrictive, the relationship: propylene resin to crosslinked product of aromatic vinyl/hydrogenated diene block copolymer (weight ratio) = 99/1 to 70/30 because

propylene resin compositions excellent in moldability and rigidity can be obtained. The production method for the propylene resin composition is optional, but includes, for example, a method of blending the propylene resin and the crosslinked product of the aromatic vinyl/hydrogenated diene block copolymer by a kneader, a roll mill, a Banbury mixer, a single screw extruder, a twin screw extruder, or the like, and a method of blending the propylene resin and the aromatic vinyl-hydrogenated diene block copolymer before subjected to crosslinking and then performing crosslinking while melt-kneading them.

[0034]

If required, the polypropylene resin composition of the present invention can be compounded with an inorganic filler such as calcium carbonate, mica, talc, silica, barium sulfate, calcium sulfate, kaolin, clay, pyrophyllite, bentonite, serisanite, zeolite, nephelincinite, attapalgite, wollastonite, ferrite, calcium silicate, magnesium carbonate, dolomite, antimony trioxide, titanium oxide, iron oxide, molybdenum disulfide, graphite, gypsum, glass beads, glass powder, glass balloons, glass fibers, quartz, quartz glass, etc., or organic or inorganic pigments. Further, if required, a crystal nucleating agent, a transparency imparting agent, an antiblocking agent, a releasing agent, an antistatic

agent, a slipping agent, an antifogging agent, a lubricant, a heat stabilizer, an ultraviolet stabilizer, a light stabilizer, a weathering stabilizer, a foaming agent, an antifungal agent, a rust-preventive agent, an ion trapping agent, a flame retardant, a flame retardant aid, etc., may be added.

[0035]

The propylene resin composition of the present invention can also be blended with another resin and a rubber without deviating from the objects of the present invention. In this embodiment, if required, a compatibilizing agent may be added as still another component. The another resin or rubber includes, for example, linear high density polyethylene, linear low density polyethylene, branched low density polyethylene, ethylene/vinyl acetate copolymer (EVA), ethylene/ethyl acrylate copolymer, poly(1-butene), poly(1-hexene), polyamide, polyester, poly(4-methyl-1-pentene), styrene-based thermoplastic elastomer, and polyolefin thermoplastic elastomer, ethylene/ α -olefin random copolymer elastomer and crosslinked products thereof, ethylene/ α -olefin/diene random copolymer elastomer, and crosslinked products thereof, natural rubber, acrylonitrile butadiene rubber, butadiene rubber, isoprene rubber, styrene butadiene rubber, silicone rubber,

polynorbornene rubber, and chloroprene rubber. The compatibilizer includes, for example, adhesive polymers such as an acid-modified polyolefin, and a saponified EVA; block or graft copolymers such as a polyolefin-polyamide graft or block copolymer.

[0036]

The propylene resin composition of the invention is molded into various kinds of products by an arbitrary molding method such as film forming, vacuum forming, pressure forming, blow molding, calendaring, profile extrusion, fiber spinning, or expansion molding.

[0037]

[Examples]

The present invention will be described in more detail with reference to the following examples, but it should be understood that the invention is not construed as being limited to those examples.

[0038]

Various measurements in Examples and Comparative Examples are described below.

[0039]

Measurement of Elongation Viscosity

Using a MELTEN Rheometer (manufactured by Toyo Seiki Seisakusho Ltd.), the change of elongational viscosity with time was measured at a strain rate of 0.1

sec^{-1} . The measurement temperature was 190°C.

[0040]

Measurement of Shear Viscosity

Using a cone-plate type viscometer (MR-500, trade name, manufactured by Rheology Co.), the change of shear viscosity with time was measured at a strain rate of 0.1 sec^{-1} . The measurement temperature was 190°C.

[0041]

Measurement of Flow Activation Energy

Dynamic viscoelasticity was measured using a cone-plate viscometer (MR-500, trade name, manufactured by Rheology Co., Ltd.) at temperatures of 190°C and 230°C, to determine flow activation energy.

[0042]

Measurement of Melt Drawing Property

A melt drawing property was evaluated by a capillary rheometer (Capillograph, trade name, manufactured by Toyo Seiki Seisakusho Ltd.). The drawable maximum take-off speed was measured under the conditions that the barrel temperature was 190°C, the inside diameter of the barrel was 9.55 mm, the L/D of the die was 2.95/1 (mm), and the descending rate of the cylinder was 10 mm/min. The diameter of the taken-off strand was measured, and the obtained value was divided by the diameter of the die, which was used as a standard of the melt drawing property.

[0043]

Measurement of Melt Tension

Melt tension when the taken-off speed was 10 m/min was measured in the measurement of melt drawing property.

[0044]

Measurement of MFR

Measurement was performed according to JIS K6758 under the condition of a temperature of 230°C and a load of 2.16 kg.

[0045]

Evaluation of Sheet Forming Property

Sheets each having a thickness of 1.0 mm were formed using a 50 φ sheet forming machine (VS 50 extruder) manufactured by Tanabe Plastics Machinery Co., Ltd. The cylinder temperature was C1: 180°C, C2: 200°C, C3: 220°C, and C4: 230°C, the die head temperature was 230°C, and a screw revolution was set to 50 rpm. The obtained sheets were subjected to vacuum forming using a pressure and vacuum forming machine (manufactured by Asano Laboratories Co., Ltd.). The heater temperature was 600°C, the heating time was 30 sec, and the mold drawing ratio was 0.75.

[0046]

Example 1

To a hydrogenated block copolymer (Dynalon D1320P, trade name, manufactured by JSR Corp. styrene: 4 mol%, a

vinyl content before hydrogenation: 65%, a number average molecular weight (in terms of polystyrene): 95,000, MFR: 3.8 g/10 min), were added α,α' -bis(t-butyl peroxy-m-isopropyl)benzene (perbutyl P, trade name, manufactured by NOF Corp.): 2500 ppm, and triaryl isocyanurate, as a crosslinking aid (TAIC, trade name, manufactured by Nippon Kasei Chemical Co., Ltd): 1500 ppm, the resulting mixture was kneaded by an internal mixer (Laboplast Mill, trade name, manufactured by Toyo Seiki Seisakusho Ltd., capacity: 100ml) for 3 min at 180°C, thereby performing thermal crosslinking.

[0047]

An MFR of the obtained crosslinked product of the hydrogenated block copolymer was 0.45 g/10 min.

[0048]

The obtained crosslinked product of the hydrogenated block copolymer and a propylene homopolymer (Chisso Polypro K1011, trade name, manufactured by Chisso Corporation, MFR: 1.0 g/10 min) were melt kneaded at a weight ratio of 1:9 in a roll mill kept at 180°C. During kneading, 10,000 ppm of a hindered phenol-based stabilizer (Irganox 1010, trade name, manufactured by Ciba-Geigy Corporation), 10,000 ppm of a phosphorus-based stabilizer (Irgaphos 168, trade name, manufactured by Ciba-Geigy Corporation) both as heat stabilizers, and 5,000 ppm of

calcium stearate as a lubricant were added thereto to obtain a polypropylene resin composition. A similar procedure was repeated several times to obtain 5 kg of a specimen.

[0049]

The ratio of the maximum values of shear viscosity to elongational viscosity, melt drawing property, melt tension, and flow activation energy of the obtained propylene resin composition were measured to evaluate moldability. The results obtained are shown in Table 1.

[0050]

Example 2

A propylene resin composition was obtained in the same manner as in Example 1 except that the amount of α,α' -bis(t-butyl peroxy-m-isopropyl)benzene was 5,000 ppm, and the amount of triaryl isocyanurate was 10,000 ppm, and evaluation of various kinds of rheology properties, as well as evaluation of molding processability were performed. The results obtained are shown in Table 1.

[0051]

Incidentally, the MFR of the crosslinked product of the hydrogenated block copolymer was 0.10 g/10 min.

[0052]

Comparative Example 1

Rheology properties and moldability of the propylene

homopolymer used in Example 1 were evaluated. The results obtained are shown in Table 1.

[0053]

Comparative Example 2

A composition was obtained in the same manner as in Example 1 except that the hydrogenated block copolymer was used without subjected to crosslinking, and rheology properties and moldability were evaluated. The results obtained are shown in Table 1.

[0054]

Comparative Example 3

Rheology properties and moldability were evaluated using a long chain branch-introduced polypropylene (PF-814, trade name, manufactured by Montel Co., MFR: 3 g/10 min). The results obtained are shown in Table 1.

[0055]

[Table 1]

	Ratio of maximum values of shear viscosity to elongation viscosity	Minimum values of the ratio of strand diameter to die diameter	Melt tension (g)	Flow activation energy (kJ/mol)	Sheet forming property
Example 1	5.8	0.066	14.0	40.2	Good
	23	0.09	16.5	40.5	Good
Comparative Example	1	3	0.065	12.0	Poor
	2	3	0.064	11.8	Poor
	3	0.44	53.0	48.3	Poor

Note) Poor: There was uneven thickness.

[0056]

[Advantage of the Invention]

According to the present invention, a propylene resin composition excellent in molding processability can be obtained.